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(54) Title: ADHESIVE FOR ESTABLISHING A CONTACT BETWEEN A DEVICE AND THE SKIN OF A MAMMAL

(57) Abstract: An adhesive for establishing a contact between a device and a mammal is disclosed. The adhesive comprises a hydrogel and a plasticizer having the general formula: R^1-(-O-R^2-)$_n$-O-R^3, wherein R^1 is H or a straight or a branched alkyl group having 1-6 carbon atoms, R^2 is, independently, a straight or a branched alkylene group having 2-6 carbon atoms, R_3 is a straight or a branched alkyl group having 1-6 carbon atoms, and n is an integer from 2 to 100. The adhesive has in preferred embodiments a longer shelf life compared to traditional adhesives.

TITLE

Adhesive for establishing a contact between a device and the skin of a mammal.

TECHNICAL FIELD

The present invention relates to an adhesive for establishing an intimate contact, particular electrical contact, between a device and the skin of a mammal.

BACKGROUND ART

Adhesives for establishing an intimate contact between a device and the skin of an mammal are used in a number of well known application, such as various types of dressings, surveillance equipment such as electrodes for monitoring the heart or brain activity, and for application of electrical signals with an aim of treating certain disorders e.g. heart failures.

Conductive adhesives may establish electrical contact between an electrode and the skin for the administration of electrical signals to the body as well as for collecting electrical signals generated in the body.

A close contact between the electrode and the skin as well as the adhesion of the electrode to the skin can be secured by application of a conductive adhesive between the electrode and the skin, optionally as an integral part of the electrode.

Adhesives for establishing contact between a device and the skin of a mammal have been addressed in a number of prior art documents.

In US 4,524,087 and 4,539,996 a conductive, adhesive gel prepared from a precursor mixture of water, a polyhydric plasticizer e.g. glycerol, sodium acrylate, a photo initiator, and a cross-linking agent, is disclosed.

A similar gel is described in US 4,554,924, the sodium acrylate being substituted with acrylic acid and KOH. Also, glycerol has been used or suggested as plasticizer in the following US patent Nos 5,868,136, 5,385,679, 5,536,446, 5,520,180, 4,848,353, 4,931,282, and 5,225,473.

US 4.699.146 describes an adhesive comprising a polymerized organic polymer wherein a polyalkylene glycol, e.g. polyethylene glycol, has been added as plasticizer. In one embodiment the adhesive is electrically conductive and can be used for attachment of an electrode to a mammalian tissue.

Likewise, in the US patent Nos 5,385,679, 5,536,446, and 5,520,180 it is suggested to use polyethylene glycol as plasticizer.

In the prior art described above, only polyhydric alcohols, e.g. glycerol and polyethylene glycol, are suggested as plasticizers in adhesives for establishing contact between a device and the skin of a mammal. Thus, the number of plasticizers available for the person skilled in the art is quite limited and adhesives with the desired properties for a specific purpose may not be obtained with in the frame of the prior art.

It is a purpose of the present invention to make available further plasticizers which will improve the possibilities of customising a particular adhesive. In one embodiment, an adhesive with improved shelf life is provided. More particular, adhesives which do not

in any significant extent suffer from reduced adhesiveness and elasticity upon aging are provided.

DISCLOSURE OF THE INVENTION

The purpose according to the present invention can be obtained by an adhesive comprising a hydrogel and a plasticizer having the general formula:

$$R^1-(-O-R^2-)_n-O-R^3$$

wherein

- R^1 is H or a straight or a branched alkyl group having 1-6 carbon atoms,
- R^2 is, independently, a straight or a branched alkylene group having 2-6 carbon atoms,
- R_3 is a straight or a branched alkyl group having 1-6 carbon atoms, and
- n is an integer from 2 to 100.

Preferably R^1 is H, methyl or ethyl, more preferred H.

The individual R^2 units in the polymer chain may independently be selected from any straight or branched alkylene with 2 to 6 carbon atoms. Preferably R^2 is ethylene or propylene, in particular ethylene.

Preferably R^3 is methyl or ethyl, particulary methyl.

In a preferred embodiment, n is from 3 to 20.

A preferred plasticizer is a monomethyl polyethylene glycol. Preferably the molecular weight is in the range of 100-10,000 Daltons, more preferred 200-6,000 Daltons. Particularly preferred plasticizers include triethylene glycol monomethylether (TEG MME), polyethylene glycol dimethylether with a average

molecular weight of 250 Daltons (PEG Dimethylether 250), polyethylene glycol monomethylether with an average molecular weight of 350, 750, and 5000 Daltons (PEG 350 MME, PEG 750 MME, and PEG 5000 MME, respectively).

The plasticizers used in the adhesive according to the invention, can be a single compound selected from compounds encompassed by the above general formula or a combination of two or more plasticizers according to the general formula. For example, an adhesive with an altered ratio of tensile strength to skin tackiness may be obtained if a combination of long chained and short chained plasticizers is used.

The hydrogel typically comprise a hydrophillic polymer. The hydrophillic polymer may for instance be selected from the group consisting of polyacrylate, copolymers comprising acrylic acid, polymethacrylate, polyacrylamide, poly(vinyl alcohol), poly(ethylene oxide), poly(ethylene imine), carboxymethylcellulose, methylcellulose, poly(acrylamide sulphonic acid), polyacrylonitril, poly(vinyl pyrrolidone), agar, dextran, dextrin, carrageenan, xanthan, and guar.

Preferred hydrophillic polymers comprise ionizable groups. It is preferred that the ionizable groups are acid groups, such as carboxylic, sulphonic or nitric groups. Preferably, the ionizable group is a carboxylic acid group.

Preferred hydrophillic polymers are polyacrylates and copolymers comprising acrylic acid or a salt thereof and one or more polymerisable comonomers. For example, the comonomers may be selected from the group comprising methacrylic acid, hydroxyethyl-methacrylic acid, 2-acrylamido-2-methyl-propanesulphonic acid, vinyl pyrrolidone; or salts thereof. The weight ratio of acrylic acid to the comonomer is preferably 1:1 to

1:0.001, more preferred in the range of 1:0.75 to 1:0.25.

The hydrophillic polymers may be cross-linked through a suitable cross-binding compound. A cross-binder generally comprise two or more functional groups which provides for the connection of the hydrophillic polymer chains. The actual used cross-binder depends on the polymer system. If the polymer system is polymerized as a free radical polymerisation, a preferred cross-binder comprises 2 or 3 unsaturated double bonds. A particular preferred compound is triethylene glycoldimetacrylate (TEGDMA). The cross-binder may, if present, be comprised in the adhesive in any suitable amounts, e.g. in an amount no higher than 5 % by weight, preferably in an amount between 0.5 and 3 % by weight.

The adhesive can contain the hydrophillic polymer and the plasticizer in any suitable proportions. However, it is preferred to use a weight ratio of hydrophillic polymer to plasticizer in the range 1:0.5 to 1:4, preferably 1:1 to 1:3.

In one embodiment, the adhesive according to the invention is electrically conductive. The conductivity of the adhesive may be obtained by including electrolytes in the hydrogel. The electrolytes used according to the invention may be any suitable electrolyte known in the art having the ability to move in the adhesive. The amount and choice of the electrolytes depend on the hydrogel used. The electrolytes may be added as an ionizable salt. Preferred ionizable salts are KCl, KBr, NaCl, $NaNO_3$, AgCl, $SnCl_2$, and NaCitrate. Usually, the amount of salt used in the adhesive according to the invention is 0.2 to 5 % by weight, preferably 0.5 to 3 % by weight.

To obtain an satisfactory electrically conductive adhesive it is preferred to use a hydrogel containing a hydrophillic polymer comprising ionicized acid groups in combination with a suitable electrolyte. To obtain the acid groups in ionic form it may be of advantage to include a pH controlling agent to the hydrogel. Preferred pH controlling agents are mineral acids or bases, e.g. HCl, HBr, KOH, and NaOH. Furthermore, a suitable compliance with the skin of the subject to receive the device may be obtained by regulating the pH of the hydrogel to a pH value non-irritating to the skin. Especially, if the electrode has to be in place for a prolonged time period, it is desirable to adjust the pH value within a range acceptable to the skin.

In certain applications, such as skin electrodes, it may be preferred to obtain a pH of the adhesive which is corrosive toward the used electrode, as described in the co-pending international patent application No. PCT/DK99/00280 to the present applicant.

By suitable selection of components in the hydrogel, water may be omitted in the adhesive. However, it is preferred that the adhesive comprise a certain amount of water because it generally lower the impedance. Preferably, the ratio of hydrophillic polymer to water is 1:0.4 to 1:2, especially 1:0.5 to 1:1.5.

It may be advantageous to include a viscosity enhancing agent in the hydrogel to obtain a more viscous adhesive. A higher viscosity may be desired for certain applications, such as disposable devices. The viscosity enhancing agent can be any known thickening agent. Examples of viscosity enhancing agents are polyacrylic acid, polyacrylate, and

polyacrylamide. Generally the polymers used as viscosity enhancers have a molecular weight of 1-5 x 10^6 Daltons. Usually, the content of viscosity enhancing agent in the adhesive is below 1 % by weight.

The adhesive according to the invention may be prepared in any suitable way. Generally, such method include *in situ* polymerisation of the monomers. Preferably the polymerisation is initiated with UV-radiation optionally using a photoinitiator. A preferred process for the preparation of the adhesive according to the invention comprises the following steps:

- providing a mixture comprising
  - acrylic acid or a monomer blend comprising acrylic acid,
  - a plasticizer having the general formula: $R^1-(-O-R^2-)_n-O-R^3$ wherein R^1, R^2, R^3, and n are as defined in the preceding part of the description,
  - a photo initiator and optionally a cross-binder, water, a salt, a pH controlling agent, and a viscosity enhancing agent,
- pouring the mixture on a suitable substrate, and
- curing the mixture with UV-radiation in the absence of oxygen.

Generally, the monomers or the blend of monomers are poured in a container that excludes any kind of light. The plasticizer is then added optionally together with demineralized water, salt, pH controlling agent, and a viscosity enhancing agent. The mixture is stirred at least until the salt is dissolved and the mixture becomes homogeneous. This mixture may be stored for a few days without substantial polymerisation.

Prior to the preparation of the adhesive, the photoinitiator and optionally a cross-binder, is added to the above mixture. Stirring is continued until a homogeneous solution is obtained. A cross-binder is generally included in the mixture to obtain a less fragile adhesive. In principle, any suitable cross-binder may be used. A preferred cross-binder is triethylene glycoldimetacrylate (TEGDMA). The photoinitiator can be any adequate compound having the ability to initiate the polymerisation. A preferred compound is 2-hydroxy-2-methyl-propionphenone sold under the trademark Darocure 1173.

The mixture containing the photoinitiator is poured on a suitable substrate. The mixture may be cast on a suitable substrate, e.g. on a release film to be removed before application or on the face of the device that point towards the body of the mammal during use. In the occasion the device is a disposable electrode, the mixture is contacted with the part of the electrode plate that faces the body during operation. Usually, the thickness of the mixture is from 0.3 to 3 mm, preferably from 0.5 to 1.5 mm. However, any suitable thickness may be used for a given application. Subsequently to the casting of the mixture, it is cured. The curing is generally effected with UV-radiation. The initiation of the curing may also be conducted using peroxides, azo-compounds, or redox-systems. However, the use of UV radiation is preferred as the curing process may be controlled to a larger extent during industrial production conditions.

The curing is preferably conducted excluding significant amount of oxygen. If oxygen is allowed to come into contact with the polymericizing mixture an unacceptable quality of adhesive is obtained,

presumably because the oxygen reacts with the monomers or uncouples the polymerisation reaction.

After the curing, the free face of the adhesive is provided with a release film or may be contacted with the device if the adhesive is formed on a release film. Immediately prior to the use of the device, the release film is removed and the device is contacted with the skin of the mammal via the adhesive.

The adhesive according to the invention can be used in connection with any device to be attached to the skin of a mammal. In one embodiment the device is adapted for electrical stimulation of the body. Electrical stimulation of the body is applied for therapeutical purposes for a variety of applications: pain control, muscle re-education of partially denervated muscles, rehabilitation of innervated muscles, control of edema, wound healing, bone healing, and the infusion of pharmacologic agents. Furthermore, electrical signals or shocks may be administrated to a patient having heart fibrillation. The electrical shock administered to a patient suffering from heart fibrillation counteracts atrial or ventricular fibrillation and, if the treatment succeeds, makes the rhythm of the heart revert to the normal mode.

Pain control effected by electrical stimulation is an alternative to the treatment with traditional pharmaceutical agents. One promising method of electrical pain control is Transcutaneous Electrical Nerve Stimulation (TENS) which comprises the delivery of current via electrodes provided on the skin of the patient to stimulate nerve fibres. Various kinds of pain can be treated with TENS, including low back pain, musculoskeletal pain, arthritic pain, obstetric pain, and postoperative pain.

TENS treatment uses the pain relief mechanism of the body's own nerve system. Two treating modes are normally applied. At a relatively high frequency of 50-100 Hz the tactile or sensoric nerves are stimulated and at a lower frequency around 1-4 Hz the muscle or motoric nerves are activated. The choice of TENS treating mode is inter alia dependent on the reason, extension, skin sensibility, and diagnosis of the pain. The best pain control may be obtained with a combination of the two modes.

In another embodiment of the invention the device collects electrical signals generated in the body. The generated signals can be monitored on a suitable monitoring device. In particular, the electrical signals of the heart may be monitored on a electrocardiogram (ECG) to monitor the operation of the heart.

The device supplied with the adhesive is preferably packed in a suitable airtight package and stored until use. Suitable storage conditions are at temperatures between the freezing point of the adhesive and approximately 40°C, preferred from 2°C to 35°C. Medical devices for emergency use and for use in remote areas may be exposed to extreme conditions, such as high temperatures. The adhesive according to the present invention do not deteriorate significantly even at temperatures above normal room temperature. As shown in the attached examples, the adhesive according to the present invention does not significantly loose adhesiveness at 40°C during a time period of 5 months.

While it is not intended to be limited to a certain theory, it is believed that the plasticizer used according to the present invention does not significantly interact with the hydrophillic polymer.

The explanation for the rapid deterioration of the prior art adhesive upon aging is believed to be that the plasticizer reacts with the hydrophillic polymer. Particularly, it is believed that the hydroxy groups of the prior art polyhydric plasticizers reacts with functional groups of the hydrophillic polymer. Functional groups attached to the hydrophillic polymer may be carboxy groups. The hydroxy groups of the plasticizer may then reacts with the carboxy groups of the hydrophillic polymer and form an ester bonding. When the plasticizer is fixed, it may no longer serve the function to lubricate in the space between the hydrophillic polymers. In consequence, the adhesiveness of the adhesive decreases upon aging, especially at elevated temperatures.

The plasticizers of the present invention only have one or none hydroxy group that may interact with functionally groups of the hydrophillic polymers in the hydrogel. It was an unexpected result of the experiments supporting the present invention, that a plasticizer with a sole hydroxy group does not result in an adhesive that looses adhesiveness upon aging.

The following examples will show the invention in further details, however the examples shall be regarded as illustratory only and are not intended to be construed limiting for the invention.

EXAMPLES

The following general procedure was used for the preparation of adhesives.

At room temperature, the following components, if present, are mixed under agitation in a dark container in the amounts indicated below: Demineralized water,

salt, acrylic acid, plasticizer, pH controlling agent, viscosity enhancing agent and optionally comonomers. The mixture is agitated, at least until the mixture is homogenous and the salt is dissolved.

Then a photoinitiator and a cross-binder is added and the stirring is continued for one hour.

The mixture is poured on a siliconized film covered with a tissue. The thickness of the mixture is 1 mm. The mixture is placed in a UV-radiation chamber in a oxygen-free atmosphere and irradiated with UV-rays until the gel is fully cured, i.e until the content of residue monomers in the produced adhesive is at or below 0,1 % by weight.

All the figures in the subsequent compositions are percentages by weight. The following abbreviations are used:

MME - monomethylether
PEG - polyethylen glycol
TEGDMA - triethyleneglycol dimetacrylate
Darocure 1173 - 2-hydroxy-2-methyl-propionphenone
TEG - triethylene glycol
PAA - polyacrylic acid
AMPS - 2-acrylamido-2-methyl-1-propanesulphonic acid
PEG 350 MME - Polyethylene glycol with one methylated end-group, the compound has a molecular weight of 350 Daltons,
PEG 750 MME - Polyethylene glycol with one methylated end-group, the compound has a molecular weight of 750 Daltons,
PEG 5000 MME - Polyethylene glycol with one methylated end-group, the compound has a molecular weight of 5000 Daltons,

MBA - Methylene bis-acrylamide
HEC - Hydroxyethyl cellulose

Example 1

Production of adhesives

Adhesives with the compositions shown in table I was produced using the above procedure.

Table I

	CONTROL (cf. US 4,554,924)	Composition 1 (Invention)
Demineralized water	4	15
potassium chloride	4,57	2,0
glycerin 87 %	70,94	0
PEG 350 MME	0	59,2
TEGDMA	0,21	1,0
Darocure 1173	0,28	0,3
Acrylic acid	20,0	22,5

Peel tests

Peel tests were conducted to evaluate the adhesiveness during storage. The two adhesives indicated in table 1 above were subjected to the following treatment: A strip of the adhesive is laminated in a width of 25 mm on a steelplate and rests for 30 minutes at room temperature. The strip is

peeled of in an angle of 90° using a pulling apparatus that measures the force used. The average force is the peel number.

The peel number is a measure for the adhesiveness of the adhesive, a high peel number is indicative for a good adhesiveness whereas a low peel number is indicative for a poor adhesiveness.

The adhesives of example 1 were aged in a airtight foil at 40°C for the various time periods indicated in table II. After the aging the peel number was measured.

Table II

Aging period	CONTROL	COMPOSITION 1
start = 0 day	5,3 N	2,2 N
½ month	2,8 N	
1 month		1,8 N
3 months	0,5 N	
5 months		2,4 N
16 months	0,1 N	

The uncertainties of these figures are estimated to 10 % of the measured value.

The peel numbers shows that the control adhesive starts with a high adhesiveness, which, however, rapidly decreases. The adhesive according to the invention has a less initial adhesiveness but remains substantially unaltered after aging for 5 months. After 3 months of aging at 40°C the control adhesive is rubber-like and can be crumbled. The gel according to the invention remained soft and maintained an acceptable adhesiveness even after 5 months at 40 °C.

Example 2

Additional formulations of adhesives

The following compositions was prepared in accordance with the procedure indicated above. "Comp." is used as an abbreviation for composition. All the compositions are transparent gels which can adhere to the skin of a human being.

	Comp. 2	Comp. 3	Comp. 4
Water	15.0	15.0	15.0
PEG Dimehytlether 250	60.19		
TEG MME		60.19	59.94
Potassium chloride	1.0	1.0	1.0
TEGDMA	1.0	1.0	1.25
Darocure 1173	0.31	0.31	0.31
Acrylic acid	22.5	22.5	22.5

	Comp. 5	Comp. 6
Water	15.0	15.0
TEG MME	57.66	60.19
Potassium chloride	1.0	1.0
TEGDMA	1.0	1.0
Darocure 1173	0.34	0.31
Hydroxyethyl methacrylate		7.5
Acrylic acid	25.0	15.0

	Comp. 7	Comp. 8	Comp.9
Water	15.0	15.0	15.0
TEG MME	60.0	58.54	57.57
Potassium chloride	1.0	2.0	2.0
TEGDMA	1.2	1.15	1.1
Darocure 1173	0.30	0.31	0.33
Acrylic acid	22.5	23.0	24.0

	Comp. 10	Comp. 11
Water	15.0	15.0
TEG MME	49.0	39.0
PEG dimethylether 250	10.0	20.0
Potassium chloride	2.0	2.0
TEGDMA	1.2	1.2
Darocure 1173	0.3	0.3
Acrylic acid	22.5	22.5

	Comp. 12	Comp. 13
Water	37.5	37.5
PEG 750 MME	37.5	
PEG 5000 MME		37.5
Potassium chloride	1.2	1.35
TEGDMA	1.0	0.85
Darocure 1173	0.3	0.3
Acrylic acid	22.5	22.5

	Comp. 14	Comp. 15	Comp. 16	Comp. 17
Water	15.0	15.0	15.0	15.0
PEG 750 MME				25.0
TEG MME	59.0	58.7	58.2	34.2
KCl	2.0	2.0	2.0	2.0
KOH		0.5	1.0	1.0
TEGDMA	1.1	1.0	1.0	1.0
Darocure 1173	0.4	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5	22.5

	Comp. 18	Comp. 19	Comp. 20
Water	15.0	15.0	15.0
PEG 5000 MME	25.0	25.0	
TEG MME	34.2	34.7	56.5
KCl	2.0	2.0	2.0
TEGDMA	1.0	0.5	1.1
Darocure 1173	0.3	0.3	0.4
Acrylic acid	22.5	22.5	25.0

	Comp. 21	Comp. 22	Comp. 23
Water	15.0	15.0	15.0
TEG MME	51.5		
PEG 350 MME		59.2	39.2
PEG 750 MME			20.0
Potassium chloride	2.0	2.0	2.0
TEGDMA	1.1	1.0	1.0
Darocure 1173	0.4	0.3	0.3
Acrylic acid	30.0	22.5	22.5

	Comp. 24	Comp. 25	Comp. 26	Comp. 27
Water	15.0	10.0	15.0	15.0
PEG 350 MME	39.2	64.2	58.2	57.2
TEG MME	20.0			
KCl	2.0	2.0	3.0	4.0
TEGDMA	1.0	1.0	1.0	1.0
Darocure 1173	0.3	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5	22.5

	Comp. 28
Water containing 1 % PAA 1,250,000	15.0
PEG 350 MME	59.2
Potassium chloride	2.0
TEGDMA	1.0
Darocure 1173	0.3
Acrylic acid	22.0

	Comp. 29	Comp. 30	Comp. 31
Water			20.0
Water with 1 % PAA $3x10^6$	15.0		
Water with 1 % PAA $4x10^6$		15.0	
PEG 350 MME	59.2	59.2	52.2
Potassium chloride	2.0	2.0	4.0
TEGDMA	1.0	1.0	1.0
Darocure 1173	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5

	Comp. 32	Comp. 33	Comp. 34
Water	15.0	15.0	
Polyacrylamide		1.0	
PEG 350 MME	57.2	58.2	74.2
Sodium nitrate	4.0		
Potassium chloride		2.0	2.0
TEGDMA	1.0	1.0	1.0
Darocure 1173	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5

	Comp. 35	Comp. 36	Comp. 37	Comp. 38
Water	20.0	25.0	30.0	35.0
PEG 350 MME	54.2	49.2	44.2	39.2
KCl	2.0	2.0	2.0	2.0
TEGDMA	1.0	1.0	1.0	1.0
Darocure 1173	0.3	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5	22.5

	Comp. 39	Comp. 40	Comp. 41	Comp. 42
Water	25.0	35.0	25.0	35.0
PEG 350 MME	47.95	37.95	47.7	37.7
KCl	4.0	4.0	4.0	4.0
TEGDMA	0.25	0.25	0.5	0.5
Darocure 1173	0.3	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5	22.5

	Comp. 43	Comp.44	Comp. 45	Comp. 46
Water	25.0	25.0	25.0	20.0
PEG 350 MME	47.7	47.7	47.7	52.45
Sodium nitrate	3.5	3.0		
KCl	0,5	1.0		4.0
Sodium citrate			4.0	
TEGDMA	0.5	0.5	0.5	0.75
Darocure 1173	0.3	0.3	0.3	0.3
Acrylic acid	22.5	22.5	22.5	22.5

	Comp. 47	Comp. 48	Comp. 49	Comp. 50
Water	20.0	15.0	15.0	15.0
PEG 350 MME	54.45	59.1	59.0	56.67
KCl	2.0	2.0	2.0	2.0
TEGDMA	0.75	1.1	1.2	1.0
Darocure 1173	0.3	0.3	0.3	0.33
Acrylic acid	22.5	22.5	22.5	25.0

	Comp. 51	Comp. 52	Comp. 53
Water	15.0	15.0	15.0
PEG 350 MME	60.2	60.2	60.2
Potassium chloride	1.0	1.0	1.0
TEGDMA	1.0	1.0	1.0
Darocure 1173	0.3	0.3	0.3
Acrylic acid	17.5	12.5	17.5
Methacrylate	5.0	10.0	
AMPS			5.0

	Comp. 54	Comp. 55
Water	15.0	15.0
PEG 350 MME	60.2	60.2
Potassium chloride	1.0	1.0
TEGDMA	1.0	1.0
Darocure 1173	0.3	0.3
Acrylic acid	17.5	12.5
Vinyl pyrrolidone	5.0	10.0

	Comp. 56
AMPS (50% aq)	58.1
PEG 350 MME	25.0
NaCl	1.0
MBA (10% aq)	0.8
Darocure 1173	0.1
Vinylpyrolidone	15.0

	Comp. 57	Comp. 58	Comp. 59
AMPS (58% aq)	80.0	80.0	80.0
NaH_2PO_4		0.3	0.25
PEG 350 MME	18.4	17.7	17.75
PAA			5.0
NaCl	1.0	1.0	1.0
MBA (10% aq)	0.8	0.90	0.90
Darocure 1173	0.1	0.10	0.10

	Comp. 60	Comp. 61	Comp. 62
AMPS (50% aq)	85.0	85.0	85.0
NaH_2PO_4		0.25	0.25
PEG 350 MME	8.1	10.95	12.05
PAA			1.0
PVP K90		2.0	
NaCl	1.0	1.0	1.0
MBA (10% aq)	0.8	0.7	0.6
Darocure 1173	0.10	0.10	0.10

	Comp. 63	Comp. 64	Comp. 65
AMPS (50% aq)	85.0	85.0	
AMPS (58% aq)			80.0
NaH_2PO_4	0.25	1.25	1.25
PEG 350 MME	12.25	11.05	17.95
PVP K-90		1.0	
HEC	1.0		
NaCl	1.0	1.0	1.0
MBA (10% aq)	0.4	0.6	0.7
Darocure 1173	0.1	0.10	0.10

	Comp. 66	Comp. 67	Comp. 68
AMPS (58% aq)	80.0	75.0	75.0
NaH_2PO_4	0.25	0.25	0.25
PEG 350 MME	18.15	22.75	22.65
NaCl	1.0	1.0	1.0
MBA (10% aq)	0.5	0.9	1.0
Darocure 1173	0.10	0.10	0.10

PATENT CLAIMS

1. An adhesive for establishing a contact between a device and the skin of a mammal, comprising a hydrogel and a plasticizer having the general formula:

$$R^1-(-O-R^2-)_n-O-R^3$$

wherein

- R^1 is H or a straight or branched alkyl group having 1-6 carbon atoms,
- R^2 is, independently, a straight or branched alkylene group having 2-6 carbon atoms,
- R_3 is a straight or branched alkyl group having 1-6 carbon atoms, and
- n is an integer from 2 to 100.

2. The adhesive according to claim 1, wherein R^1 is H.

3. The adhesive according to claim 1 or 2, wherein R^2 is ethylene or propylene.

4. The adhesive according to any of the claims 1 to 3, wherein R^3 is methyl or ethyl.

5. The adhesive according to any of the claims 1 to 4, wherein n is from 3 to 20.

6. The adhesive according to any of the preceding claims, wherein the plasticizer is a monomethyl polyethylene glycol with a molecular weight in the range of 100-10,000 Daltons, preferably 200-6,000 Daltons.

7. The adhesive according to any of the claims 1 to 6, which is electrically conductive.

8. The adhesive according to any of the claims 1 to 7, wherein the hydrogel comprises a hydrophillic polymer selected from the group comprising polyacrylic acid and copolymers comprising acrylic acid and one or

more comonomers selected from the group comprising methacrylic acid, hydroxyethyl-methacrylic acid, 2-acrylamido-2-methyl-propanesulphonic acid, and vinyl pyrrolidone, or salts thereof; the weight ratio of acrylic acid to the comonomer being 1:1 to 1:0.001, preferably in the range of 1:0.75 to 1:0.25.

9. The adhesive according to any of the preceding claims wherein the weight ratio of hydrophillic polymer to plasticizer is 1:0.5 to 1:4, preferably 1:1 to 1:3.

10. The adhesive according to any of the preceding claims, wherein the hydrogel comprises water, the ratio of hydrophillic polymer to water preferably being 1:0.4 to 1:2, especially 1:0.5 to 1:1.5.

11. The adhesive according to any of the preceding claims which comprises a viscosity enhancing agent, preferably polyacrylic acid having a molecular weight of 1 - 5 x 10^6 Daltons.

12. A process for the production of an adhesive comprising the steps of:

- providing a mixture comprising
  - acrylic acid or a monomer blend comprising acrylic acid,
  - a plasticizer having the general formula: $R^1-(-O-R^2-)_n-O-R^3$ wherein R^1, R^2, R^3, and n are as defined in the claims 1 to 5,
  - a photoinitiator and optionally a cross-binder, water, a salt, a pH controlling agent, and a viscosity enhancing agent,
- pouring the mixture on a suitable substrate,
- curing the mixture with UV-radiation in the absence of oxygen.

13. Process for the production of an adhesive according to claim 12, wherein the plasticizer is a monomethyl polyethylene glycol with a molecular weight in the range of 100-10,000 Daltons, preferably 200-5,000 Daltons.

14. Process for the production of an adhesive according to claim 12 or 13, wherein the monomer blend comprising acrylic acid further comprises one or more comonomers selected from: methacrylic acid, hydroxyethyl-methacrylic acid, 2-acrylamido-2-methyl-propanesulphonic acid, vinyl pyrrolidone or salts thereof.

15. Process for the production of an adhesive according to any of the claims 12-14, wherein the mixture to be polymerized in addition as viscosity enhancing agent comprises polyacrylamide or polyacrylic acid with a molecular weight of 1-5 x 10^6 Daltons.

16. Adhesive obtainable according to any of the claims 12-15.

17. Use of an adhesive according any of the claims 1-11 or claim 16 for adhering an electrode to the skin of a mammal.

18. An electrode comprising the adhesive according to claims 1-11 or claim 15.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/DK 00/00507

A. CLASSIFICATION OF SUBJECT MATTER

IPC7: A61L 24/04, C09J 9/02, C09J 133/00
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC7: A61B, A61L, A61N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5536768 A (STEVEN S. KANTNER ET AL), 16 July 1996 (16.07.96), column 2, line 62 - column 3, line 23; column 5, line 41 - line 64; column 7, line 1 - line 39, column 9, line 60 - column 10, line 2; ex. 10 and 14; abstract --	1-18
X	WO 9724149 A1 (MINNESOTA MINING AND MANUFACTURING COMPANY), 10 July 1997 (10.07.97), page 1, line 6 - line 13; page 7; page 9, line 3 - line 14, page 13, line 26 - page 14, line 2; claims 1-5,15 --	1-18
X	US 5433892 A (ZBIGNIEW CZECH), 18 July 1995 (18.07.95), claims 1 and 14 --	1-18

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier application or patent but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
28 November 2000	09. 02. 2001
Name and mailing address of the International Searching Authority European Patent Office P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel(+31-70)340-2040, Tx 31 651 epo nl, Fax(+31-70)340-3016	Authorized officer BARBRO NILSSON/Els Telephone No.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/DK 00/00507

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	Electrochimica Acta, Volume 40, No 13-14, 1995, Jiazeng Sun et al, "Poly Methacrylate-Plasticiser-Salt Blends as Solid Polymer Electrolytes", page 2301 - page 2304, see especially abstract --- --------	1-18

INTERNATIONAL SEARCH REPORT

Information on patent family members

02/11/00

International application No.
PCT/DK 00/00507

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